



Best Available Copy Sheet 1 of 1

FORM PTO-	Atty. Docket No.: C382.12-0146	Appl. No.: 10/783,682
LIST OF PATENTS AND PUBLICATIONS FOR APPLICANT'S INFORMATION DISCLOSURE STATEMENT	First Named Inventor: Harvey A. Restaino et al.	
	Filing Date February 20, 2004	Group Art: 2838

U.S. PATENT DOCUMENTS

Examiner Initial	Document No.	Date	Name	Class	Sub Class	Filing Date If Appropriate
SB	AA	5,357,519	10/1994	Martin et al.	371	15.1
	AB	6,016,047	1/2000	Notten et al.	320	137
	AC	6,449,726	9/2002	Smith	713	340
	AD	6,215,275	4/2001	Bean	320	106
	AE	6,100,670	8/2000	Levesque	320	150
	AF	6,271,643	8/2001	Becker et al.	320	112
	AG					
	AH					
	AI					
	AJ					

FOREIGN PATENT DOCUMENTS

	Document No.	Date	Country	Class	Sub Class	Translation Yes No
SB	AK	GB 2 387 235 A	08.10.2003	United Kingdom		
SB	AL	WO 00/16083	23.03.00	WIPO		
SB	AM	WO 01/16614	08.03.2001	WIPO		
SB	AN	WO 01/16615	08.03.2001	WIPO		
	AO					

OTHER ART (Including Author, Title, Date, Pertinent Pages, Etc.)

AP	Search Report from Application No. GB0421447.4
AQ	Examination Report from Application No. GB0417678.0
AR	Champlin et al., K. S., "Results of Discrete Frequency Immittance Spectroscopy (DFIS) Measurements of Lead Acid Batteries", INTELEC 2001, October 2001.

EXAMINER:

DATE CONSIDERED:

EXAMINER: Initial if citation considered, whether or not citation is in conformance with MPEP 609; draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.



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Your Reference: REC/NPT/P76032GB00
Application No: GB0421447.4

28 January 2005

Dear Sirs

Patents Act 1977: Search Report under Section 17(5)

I enclose two copies of my search report and two copies of the citations. The enclosed citations are examples of many similar documents.

Other search results

If you have applied to another patent office for a patent for this invention you will be receiving from them the results of their search. If you decide to proceed with the present application you are asked to provide me with a copy of any such official search report; or to email identifying details of the cited documents (including any category assigned in the report) to the email address above.

Cut-off date This request applies to search reports that you have received before the date when you send a response to our first examination report under section 18(3) or section 18(4); if you make no response to an initial section 18(4) report the cut-off date is two months after the date of that report. Tell us about a search report sooner rather than later if that would allow it to be considered during our first examination.

Exceptions You do not have to supply details of a search report that (1) shows a nil response, or (2) has been published by WIPO or EPO, or (3) you have already supplied to us on a previous GB application.

Publication

I estimate that, provided you have met all the formal requirements, preparations for publication of your application will be completed soon after 1 March 2005. At this time you will receive a letter confirming the exact date when the preparations for publication will be



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Application No : GB0421447.4

Page 2

28 January 2005

completed. This letter will also tell you the publication number and date of publication of your application.

Withdrawal/amendment

If you wish to withdraw your application before it is published you must do so before the preparations for publication are complete. **WARNING** – after preparations for publication are complete it will NOT be possible to withdraw your application from publication.

If you wish to file amended claims for inclusion with the published application you must do so before the preparations for publication are completed. If you write to the Office less than 3 weeks before 1 March 2005 please mark your letter prominently:

"URGENT - PUBLICATION IMMINENT".

Yours faithfully

Mr Stuart Jarvis
Examiner



INVESTOR IN PEOPLE

Application No: GB0421447.4

Examiner: Mr Stuart Jarvis

Claims searched: 1-57

Date of search: 27 January 2005

Patents Act 1977: Search Report under Section 17

Documents considered to be relevant:

Category	Relevant to claims	Identity of document and passage or figure of particular relevance
X	1-55	US6215275 B1 Bean - See figure 4 and col 4 lines 31-63
X	1-55	US5831435 A Troy - See figure 3, col 2 lines 39-49 and col 4 lines 45-52
X	1-55	US6016047 A Notten - See col 10 lines 37-49
X	1-55	US6449726 B1 Smith - See col 6 lines 21-29
X,P	1-55	GB2387235 A Schinner - See page 11 lines 3-9
X,P	1-55	US2003/0184262 A1 Makhija - See figure 1, paragraph 17 and claims 1 and 2

Categories:

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.

Field of Search:

Search of GB, EP, WO & US patent documents classified in the following areas of the UKC^X:

G1U

Worldwide search of patent documents classified in the following areas of the IPC⁰⁷

G01R

The following online and other databases have been used in the preparation of this search report

EPODOC, WPI, JAPIO



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Your ref : P73416GB00
Application No: GB0417678.0
Applicant : Keith S Champlin

Examiner : Eleanor Hogan
Tel : 01633 814798
Date of report : 24 January 2005

Latest date for reply: 24 May 2005

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Patents Act 1977 Examination Report under Section 18(3)

Basis of the examination

1. I have examined your application in the form that it was printed by WIPO when it was in the international phase.

Novelty (Section 1(1)(a))

2. The invention as defined in claims 1-3, 5, 6, 10 and 11 is not new because it has already been disclosed in each of the following documents:

K S Champlin & K Bertness, "Proceedings of 23rd International Teleco Conference (INTELEC)", published October 2001, IEE, pp 433-440, "Results of discrete frequency immittance spectroscopy (DFIS) measurements of lead acid batteries".

WO 01/16615 A1 (CHAMPLIN)

See abstract and figs. 1 & 2; Relevant to claims 1-3.

WO 01/16614 A1 (CHAMPLIN)

See abstract and figs. 1 & 2; Relevant to claims 1 & 2.

WO 00/16083 A1 (CHAMPLIN)

See abstract and figs. 2 & 7; Relevant to claims 1 & 2.

3. The above CHAMPLIN & BERTNESS paper shows the use of an equivalent circuit comprising a series combination of a series R-L subcircuit and a parallel G-C subcircuit to monitor various battery parameters including SCO, maximum exchange current, maximum double layer capacitance. Therefore claims 1-3, 5, 6, 10 and 11 are anticipated.

4. All the above patent documents disclose the determination of battery parameters by applying time varying electrical excitation to the battery, measuring a time varying electrical response and using this response to determine the values of circuit elements in an equivalent circuit. The equivalent circuits comprise a series combination of a subcircuit containing a resistive element and an inductive element, and two subcircuits each containing a resistive element and capacitive element.

5. All these circuits include resistors that are labelled by either their conductance (G) or resistance (R). Given that G is merely the reciprocal of R, these documents anticipate the circuit of the application in suit. Hence claims 1 and 2 are not novel.



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[Examination Report contd.]

6. Furthermore, state of charge (SOC) is determined WO '615 and so claim 3 is not new either.

7. Please note that the above documents were found during the supplementary search at examination stage. WO '615 is the equivalent of US6313607 B which was cited on the US equivalent of the application in suit (US 6737831 B).

Inventive step (Section 1(1)(b))

8. The invention as defined in claims 4 & 7-11 is obvious in view of what has already been disclosed in the following documents:

See novelty citations above.

9. Although the cited documents do not show the calculation of all the specific parameters mentioned in the claims, it appears that a person skilled in the art would find it obvious either to use the method described in the documents in order to calculate the specified parameters or to derive these parameters from those already calculated. There appears to be nothing inventive in the extension of the disclosures of the above documents to the determination of other parameters, and unless you can argue otherwise, claims 4 and 7-11 lack the required inventive step.

Clarity, Support and Scope of Invention (Sections 14(5)(b)&(c))

10. The scope of the invention is not entirely clear because the "summary of invention" section on page 6 is not entirely consistent with the features set out in the independent claims. Amendment is required to ensure any purported statements of invention are consistent with the main claims, ensuring that the description does not suggest any features that not present in the main claims are essential (for example page 4 suggests that one of the display, alarm and control features is essential but none of these features are present in the claims).

11. Furthermore the scope of the invention is obscure by the final sentence of page 28 referring to "spirit and scope of invention". This sentence should therefore be removed.

12. Claims 1, 2, 10 and 11 do not make it entirely clear that it is the apparatus (claims 1 and 10) or method (claims 2 and 11) and not the battery that is "comprising" the stated features.

13. Claims 1 and 10 initially state that the parameters are "determined" but later state that the computations circuitry is adapted to "evaluate" the parameters. It appears that



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Application No : GB0417678.0

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[Examination Report contd.]

evaluate is intended to have the same meaning as *determine* (indeed claims 2 and 11 use "determining") and so consistent terminology should be used throughout the claims.

Field of search

14. The field of search was extended to include the ECLA classification term applied to this case by the EPO, namely G01R 31/36V1A.

RESULTS OF DISCRETE FREQUENCY IMMITTANCE SPECTROSCOPY (DFIS) MEASUREMENTS OF LEAD ACID BATTERIES

K S Champlin and K Bertness

University of Minnesota, U.S.A. and
Midtronics, Inc., U.S.A.

Electrochemical Impedance Spectroscopy (EIS) has become a common research tool in laboratory studies of many electrochemical processes. The EIS methodology begins with an assumed equivalent-circuit model of the system under investigation. It then utilizes either a graphical analysis procedure or a nonlinear complex least-squares curve-fitting procedure to make the assumed model conform to measurements of the system's complex impedance acquired over a wide range of frequencies. (See, e.g., Macdonald (1), Robinson (2).) Although a few EIS investigations of lead-acid battery systems have been reported, very little work has been reported on *real* batteries – batteries actually found in the field (Karden et al (3), Huet (4)). In addition, most EIS battery investigations have focused on the millihertz and microhertz frequency range. At such low frequencies, a single impedance measurement can take hours. The battery's properties may change in the meantime. The combination of the graphical or least-squares curve-fitting procedure, the many measurements required, and the low frequencies employed, means that conventional EIS takes a very long time. Such a time-consuming procedure is probably not very practical for testing real batteries in the field.

Discrete Frequency Immittance Spectroscopy (DFIS) also begins with a circuit model of the battery and uses small-signal measurements of complex immittance (i.e., either impedance or its reciprocal, admittance). However, instead of measuring the complete immittance spectrum, measurements are made at only a few, judiciously chosen, discrete frequencies. A *direct* calculation then yields element values of the circuit model *exactly* in terms of the measurements. Although there is no reason why DFIS could not be employed at millihertz or microhertz frequencies, we have found very significant information in the spectrum above one hertz. The combination of using $f > 1$, making few measurements, and doing the analysis with a *direct* calculation, means that DFIS can be performed very quickly – generally in less than one minute.

REVIEW OF DFIS PRINCIPLES

The DFIS methodology (Champlin and Bertness (5)) employs the $2n$ -element circuit model of Figure 1 to represent the battery. This model comprises a dc voltage source in series with a single series R-L subcircuit and a series array of $(n-1)$ parallel G-C subcircuits. The model of Figure 1 simply describes a

generic frequency-dependent circuit with no attempt made to initially identify any individual elements with particular processes occurring within the battery.

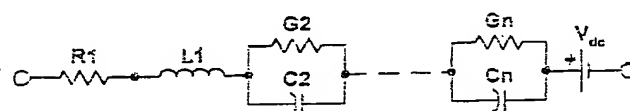


Figure 1. Generic DFIS circuit model.

The elements of the generic circuit model are evaluated with an *exact* calculation that determines their $2n$ values from the real and imaginary parts of the complex immittance measured at n discrete frequencies. Measurements at two frequencies will evaluate a 4-element circuit; three frequencies, a 6-element circuit, and so on.

Figure 2 shows DFIS results for a typical 12V automotive SLI battery using $n = 2$: $f_1 = 5$ Hz, and $f_2 = 1000$ Hz. Two widely separated time constants, both much smaller than one second, are noted.

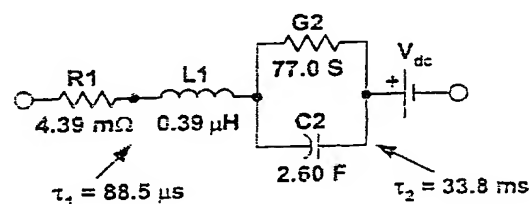


Figure 2. DFIS model evaluation for $n = 2$.

As a check on this technique, the complex admittance of the circuit model of Figure 2 was calculated as a function of frequency and plotted along with the battery's measured complex admittance spectrum for comparison. These results are shown in Figure 3.

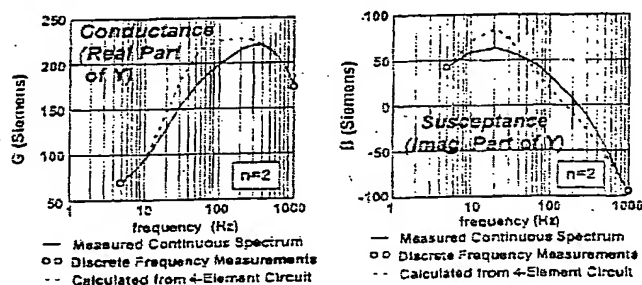


Figure 3. Measured $Y = G + jB$ spectrum compared with that calculated from model of Figure 2.

One sees that the measured and calculated values agree exactly at the two discrete DFIS frequencies. However, away from these frequencies the agreement is poor. This indicates that the model of Figure 2 does not adequately represent the battery's small-signal ac response over this frequency range.

Figure 4 shows DFIS results for the same battery using $n = 3$: $f_1 = 5$ Hz, $f_2 = 70$ Hz, and $f_3 = 1000$ Hz. Three widely separated time constants are now noted. All three are much smaller than one second.

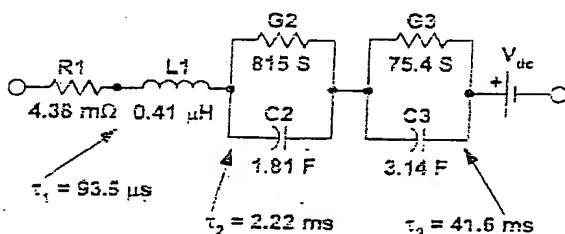


Figure 4. DFIS model evaluation for $n = 3$.

The complex admittance of the model of Figure 4 was calculated as a function of frequency and plotted along with the measured complex admittance spectrum for comparison. This comparison is shown in Figure 5.

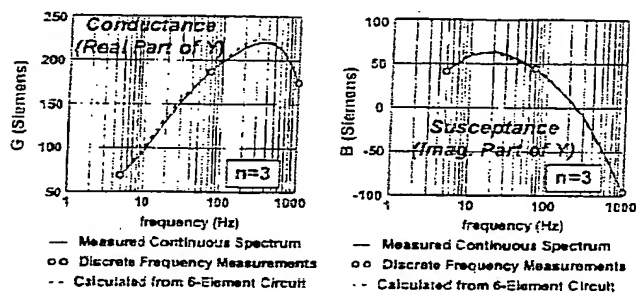


Figure 5. Measured $Y = G + jB$ spectrum compared with that calculated from model of Figure 4.

Once again measured and calculated values agree exactly at the DFIS frequencies. Away from these frequencies however, the agreement is now also very good. Such agreement proves the model of Figure 4 to be an excellent small-signal representation of the battery over the complete frequency range from 5 Hz to 1000 Hz. Accordingly, the six element values disclosed in Figure 4 contain virtually the same information as the complete spectral plots of Figure 5. However, they embody this information in a far more concise form that is much easier to store, analyze, and manipulate than the spectral plots from which they were derived.

PHYSICAL INTERPRETATION

By analyzing three-frequency DFIS measurements of many lead-acid batteries, a consistent picture has emerged. Resistance R_1 is believed to describe the current-induced drop in electric potential across the metal connectors, plates, separators, and electrolyte — with the majority of R_1 associated with the electrolyte. In this interpretation, conductance $G_1 = 1/R_1$ is directly related to high-rate load-test current such as CCA. Inductance L_1 appears to simply describe energy storage in the battery's magnetic field, with very little electrochemical component to L_1 yet observed.

Both the G_2 - C_2 subcircuit and the G_3 - C_3 subcircuit are believed to be linearized representations of nonlinear electrochemical processes. Because of their small time constants, both processes are believed to occur at the negative electrode surfaces. Why the negative electrode surfaces? The answer can be seen in Figure 6.

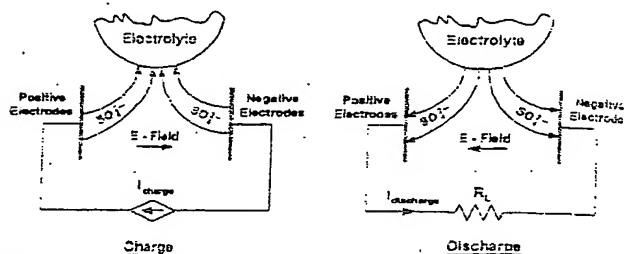


Figure 6. Directions of E-field and SO_4^{2-} ion flow for battery charge and for battery discharge.

The SO_4^{2-} ions, which are involved in reactions at both positive and negative electrode surfaces, flow from both electrodes during charging and to both electrodes during discharging. As seen in Figure 6, the electric field aids migration of these (negative) ions to/from the negative electrodes but opposes it to/from the positive electrodes. Accordingly, the rate of electrochemical reactions at the positive electrode surfaces is controlled by diffusion — a process that requires the existence of concentration gradients. Concentration gradients take time to build up. Positive-electrode reactions are therefore orders of magnitude slower than the field-assisted reactions at the negative-electrode surfaces.

This conclusion may explain why Shukla et al (6) found time constants approximately 10^4 times longer than our observed τ_2 and τ_3 for a circuit model similar to Figure 4 that they obtained by analyzing the long-term transient response of a lead-acid battery. They were quite possibly observing positive-electrode reactions only. Similarly, results of EIS experiments on lead-acid batteries at millihertz and microhertz frequencies are probably reflections of positive-electrode behavior.

As stated above, the G_2 - C_2 subcircuit and the G_3 - C_3 subcircuit in series are believed to describe electrochemical processes at the negative electrode surfaces. These two subcircuits together are believed to describe the response of the negative electrodes to changes in an applied electrochemical potential. (Note: In the following discussion we will consider a change in electrochemical potential as an applied force and the resulting current change as the response. However, our DFIS experiments actually reverse the roles of voltage and current. In the linear regime, these two points of view are equivalent. See, e. g., reference (1), p. 6.)

Figure 7 displays graphically what an electrochemical cell or battery actually does. Faradaic processes within the battery raise the Fermi level of electrons in the metallic negative terminal above that of electrons in the

metallic positive terminal. The battery's open-circuit voltage is simply proportional to this Fermi level difference. Increasing the Fermi level difference by applying an electrical overpotential drives current into the battery. Reducing it draws current out.

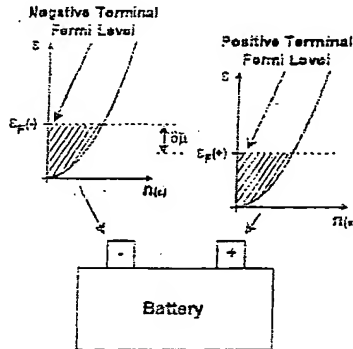


Figure 7. Total electrochemical potential difference between positive and negative battery terminals.

One can show from statistical mechanics and thermodynamics that Fermi level and electrochemical potential are equivalent. (Reference (7), p. 633.) Consequently, the Fermi level difference at the terminals is the sum total of all of the electrochemical potential differences at all of the series-connected positive and negative electrodes within the battery.

Electrode electrochemical potential differences consist of two parts: an *electrical* potential difference and a *chemical* potential difference. (See reference (7), pp. 60, 633.) The *electrical* part arises from electrical forces on charged particles, and the *chemical* part arises from concentration differences. For reasons that will be made clear below, we believe that the faster G2-C2 subcircuit responds to the *electrical* part of an electrode electrochemical overpotential and the slower G3-C3 subcircuit responds to the *chemical* part. These two subcircuits add in *series* in the circuit model of Figure 4 because the *electrical* overpotential and the *chemical* overpotential add together to form the *electrochemical* overpotential. (See, e.g., reference (7), equation (14.5.4).) Voltages add in series.

DFIS ANALYSIS OF A VRLA BATTERY

Figures 8, 9, and 10 display the results of DFIS measurements performed at 5, 50, and 500 Hz on a 12V VRLA/AGM battery of spiral-grid construction. This battery, which had seen some service, was rated 700 CCA, 95 minutes reserve capacity, and 50 Ah @ 20 hr rate. The battery was initially charged and was then repeatedly discharged at an approximate 8-hr rate in increments of approximately 3 Ah. The battery was permitted to rest for 2.5 hours after each discharge period, and the DFIS measurements were then performed. Each complete DFIS measurement passed a 1-amp p-p square wave current through the battery in the discharge direction for approximately one minute.

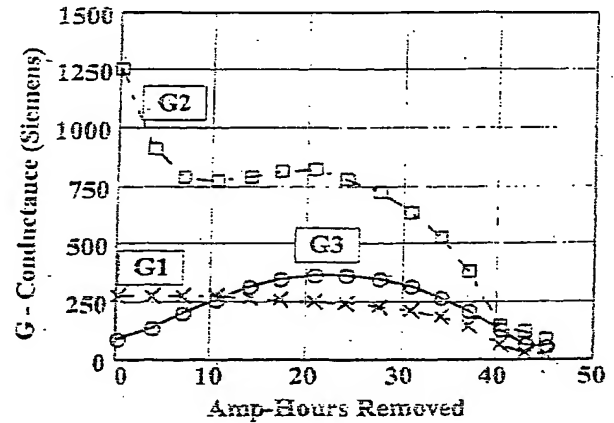


Figure 8. Conductances vs. Ah removed.

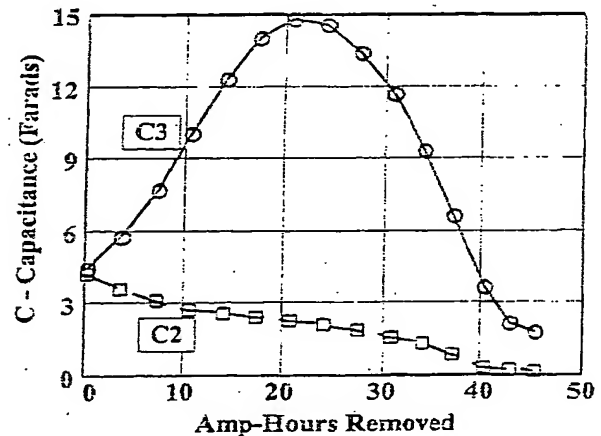


Figure 9. Capacitances vs. Ah removed.

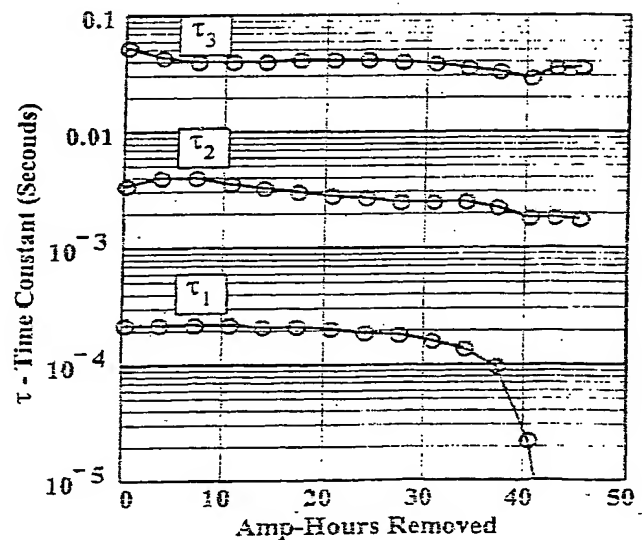


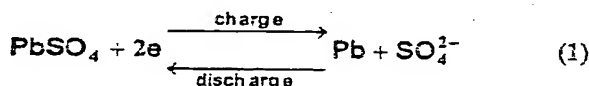
Figure 10. Time Constants vs. Ah removed.

One sees from Figure 10 that the three time constants defined in Figure 4 are again widely separated and remain so as the charge is removed.

Variation of G3 with Amp-hours Removed

Consider Figure 8. Of particular interest is the fact that G3 becomes small at full charge as well as at full discharge. This remarkable behavior sets G3 apart from G1 and G2, both of which reach *maxima* at full charge. (It is interesting to note that Shukla et al (6) observed similar behavior for one of the resistive elements in the model they derived from long-term transient response.) Such unusual behavior can be explained by assuming that G3 describes *faradaic* processes occurring at the negative electrode surfaces and considering the kinetics of such processes.

Faradaic processes at the negative electrode surfaces are of the form



in which the right-pointing arrow describes the reduction ("charge") reaction and the left-pointing arrow describes the oxidation ("discharge") reaction. The Butler-Volmer equation for this process can be written (See reference (7), equation (3.5.11).):

$$i = i_f - i_b = i_0 \left[e^{\alpha n f V_3} - e^{-(1-\alpha) n f V_3} \right] \quad (2)$$

In equation (2), i is the current in the external circuit (assumed positive in the "charge" direction); i_f and i_b are "forward" and "backward" reaction currents, respectively; i_0 is the "exchange current"; α is a "charge transfer coefficient"; n is the number of electrons (two) transferred per molecule reactant; and $f = (F/RT)$, with F being Faraday's constant, R the universal gas constant, and T the absolute (Kelvin) temperature. According to our present understanding, voltage V_3 is the *chemical* component of the *electrochemical overpotential* (positive for "charge" and negative for "discharge") at the negative electrode surfaces. (Reference (7) refers to this quantity as simply the "overpotential" $-\eta$).

The *chemical overpotential*, V_3 , drives the two reaction currents. Consider Figure 11. Under charging conditions ($V_3 > 0$), the forward ionic current, $i_f = i_0 \exp(\alpha n f V_3)$, is larger than i_0 ; while the backward ionic current, $i_b = i_0 \exp(-\{1-\alpha\} n f V_3)$ is less than i_0 . Accordingly, their difference yields a

positive ("charging") current in the external circuit. Under discharging conditions, the opposite is true. In equilibrium ($V_3 = 0$), both forward and backward ionic currents are equal to the exchange current i_0 , and the current in the external circuit is zero. Note that even with $i = 0$, there are still two equal ionic "exchange" currents flowing in opposite directions in the electrolyte. The electrolyte is continuously active.

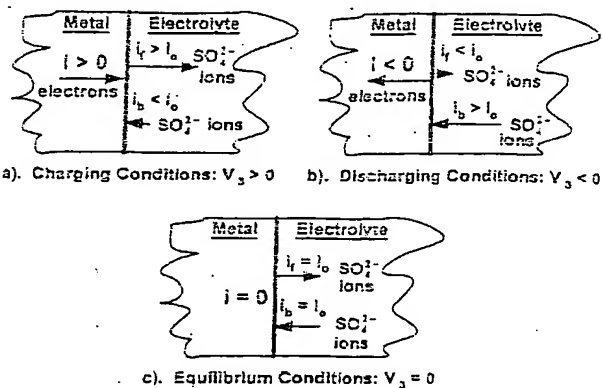


Figure 11. Conditions at negative electrode interface according to Equation (2).

The measured small-signal dynamic conductance at equilibrium, G3, follows by differentiating equation (2) with respect to $(N_C V_3)$, where N_C is the number of battery cells in series, and evaluating this derivative at $V_3 = 0$. The result is

$$G3 = \left. \frac{di}{d(N_C V_3)} \right|_{V_3=0} = \frac{n f i_0}{N_C} = (n F / N_C R T) \cdot i_0 \quad (3)$$

One sees that at any given temperature, conductance G3 is simply proportional to the exchange current, i_0 .

The observed variation of G3 with battery charge can now be *qualitatively* explained. As the battery approaches full charge, the forward, or "charge", component of the negative-electrode reaction current goes to zero since the number of sites available for the charge reaction (PbSO_4 sites) goes to zero. Because forward and backward ionic currents must be equal in equilibrium, the equilibrium backward current must likewise go to zero. Similarly, as the battery approaches full discharge, the backward, or "discharge", component of the reaction current goes to zero since the number of sites available for the discharge reaction (Pb sites) approaches zero. Again, since backward and forward ionic currents must be equal in equilibrium, the equilibrium forward current likewise goes to zero. Accordingly, the exchange current i_0 goes to zero in both cases. From equation (3), conductance G3 -- which is simply proportional to i_0 -- likewise approaches zero at both full charge and full discharge.

Equation (3) can be inverted to yield the exchange current i_0 in terms of the measured conductance $G3$. For a 12V (6-cell) battery at room temperature ($T = 293^\circ \text{K}$), equation (3) yields

$$i_0 = (N_c RT / nF) \cdot G3 = (.07574) \cdot G3 \text{ amperes} \quad (4)$$

Figure 12 displays i_0 calculated from equation (4) using the experimental $G3$ data of Figure 8. One sees that i_0 does indeed approach zero at both full charge and full discharge and reaches a maximum value of $i_{0\text{max}} = 27.5$ amperes at approximately 50 % state-of-charge.

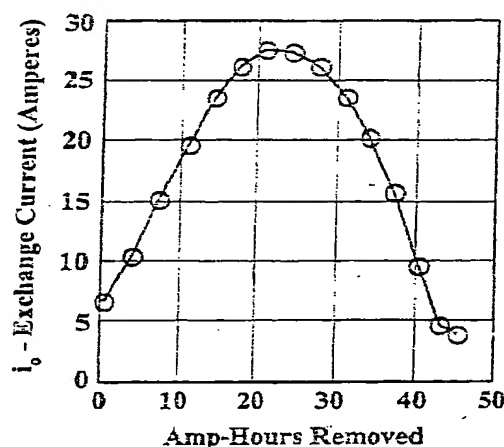


Figure 12. Negative-electrode exchange current as determined from DFIS measurements of $G3$.

By reasoning in accordance with the qualitative mechanism described above, one can write $G3$ at any particular temperature as

$$G3 = 4 \cdot G3_{\text{max}} \cdot (N_{\text{Pb}}^- / N^-) \cdot (N_{\text{PbSO}_4}^- / N^-) \quad (5)$$

where $N^- = N_{\text{Pb}}^- + N_{\text{PbSO}_4}^-$ is the total number of negative-electrode reaction sites, N_{Pb}^- is the number of such sites occupied by Pb molecules, and $N_{\text{PbSO}_4}^-$ is the number occupied by PbSO_4 molecules. The factor 4 arises because $G3$ must equal $G3_{\text{max}}$ when $N_{\text{Pb}}^- = N_{\text{PbSO}_4}^- = N^- / 2$.

We now define the "negative-electrode state-of-charge" SOC^- as the fraction of negative-electrode reaction sites that are occupied by Pb molecules. That is

$$\text{SOC}^- \equiv (N_{\text{Pb}}^- / N^-) = 1 - (N_{\text{PbSO}_4}^- / N^-) \quad (6)$$

Equation (5) can therefore be written as

$$G3 = 4 \cdot G3_{\text{max}} \cdot (\text{SOC}^-) \cdot (1 - \text{SOC}^-) \quad (7)$$

which obviously approaches zero when SOC^- approaches both unity and zero. Since i_0 is proportional to $G3$ at any particular temperature, the exchange current can likewise be written

$$i_0 = 4 \cdot i_{0\text{max}} \cdot (\text{SOC}^-) \cdot (1 - \text{SOC}^-) \quad (8)$$

Equation (7) can be rearranged into a quadratic equation for SOC^- in terms of $(G3/G3_{\text{max}})$. The solution to this quadratic is

$$\text{SOC}^- = 0.5 \pm 0.5 \sqrt{1 - (G3/G3_{\text{max}})} \quad (9)$$

in which the positive and negative roots correspond to $\text{SOC}^- > 0.5$ and $\text{SOC}^- < 0.5$, respectively.

Figure 13 displays SOC^- (in percent) calculated from equation (9) using the experimental $G3$ data of Figure 8. The state-of-charge of the negative electrodes determined in this manner varies from a maximum value of 93.6 %, with zero amp-hours removed from the battery, to a minimum value of 3.6 % after 45.2 amp-hours have been removed.

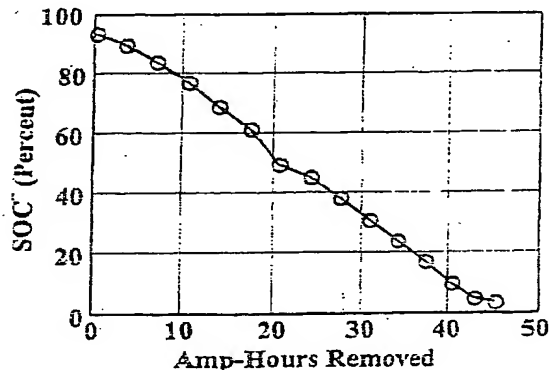


Figure 13. Negative-electrode state-of-charge as determined from DFIS measurements of $G3$.

The observed, nearly linear, variation of SOC^- with amp-hours removed provides excellent corroboration of the theory presented above.

Variation of $C2$ and $C3$ with Amp-hours Removed.

Consider Figure 9. Capacitance $C3$ is seen to mimic the behavior of $G3$ over the complete range of Ah removed. This variation can be explained by noting that $C3 = \tau_3 \cdot G3$, and that τ_3 varies very little as charge is removed from the battery (see Figure 10). The time constant τ_3 is believed to be closely related to the reaction time of the faradaic processes at the negative electrode surfaces.

Capacitance $C2$ is seen to vary in an entirely different manner as charge is removed. Instead of increasing and then decreasing, $C2$ has its largest value at "full charge" and simply decreases monotonically as charge is removed from the battery. This behavior leads us to believe that the $G2$ - $C2$ subcircuit is responsive to the nonfaradaic or electrical component of the electrochemical overpotential at the negative-electrode surfaces. In this interpretation, capacitance $C2$ represents the negative-electrode's ionic double layer capacitance. (See e.g., reference (7), pp. 6-14.)

An ionic double layer can only form on the metallic Pb molecules. It cannot form on nonconducting $PbSO_4$ molecules. It follows that $C2$ should be proportional to (N_{Pb}^-/N^-) , the fraction of the total reaction sites that are occupied by Pb molecules. Accordingly, capacitance $C2$ can be written

$$C2 = C2_{100} \cdot SOC^- \quad (10)$$

where SOC^- is the negative-electrode state-of-charge defined in equation (6), and $C2_{100}$ is the value of $C2$ that would occur at $SOC^- = 100\%$. Accordingly

$$SOC^- = (C2/C2_{100}) \quad (11)$$

Figure 14 shows a plot of SOC^- as determined from equation (11) and the experimental $C2$ data of Figure 9 by assuming that $SOC^- = 93.6\%$ with zero Ah removed. Although the plot of Figure 14 is not quite as linear as that of Figure 13, it is still sufficiently linear to lend considerable credibility to the proposed theory.

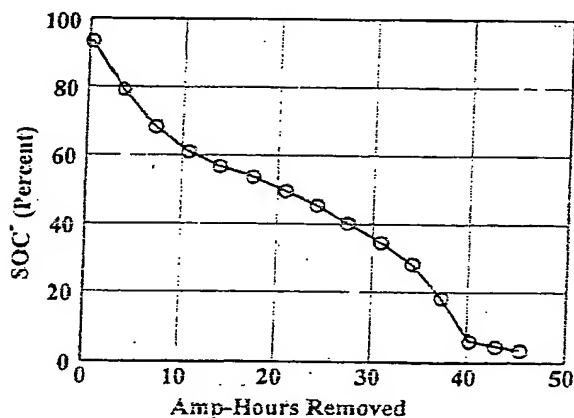


Figure 14. Negative electrode state-of-charge as determined from DFIS measurements of $C2$.

In addition to equations (9) and (11) for SOC^- , one can combine equations (7) and (10) to obtain a third expression for SOC^- . The result is

$$SOC^- = 1 - K \cdot (G3/C2) \quad (12)$$

where K is a constant defined by

$$K = (C2_{100}/4 \cdot G3_{max}) \quad (13)$$

Equation (12) is plotted in Figure 15.

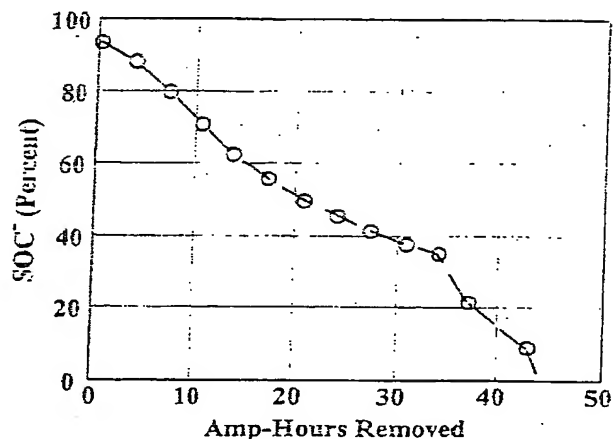


Figure 15. Negative electrode state-of-charge determined by combining measured $C2$ and $G3$.

All three equations for SOC^- agree rather well except near full-depletion. (Data points at 40 and 45 Ah-removed were excised from Figure 15 because they yielded negative values of SOC^-). However there are two advantages to using equation (12), as opposed to using equations (9) and (11), to determine SOC^- from experimental data. First, equation (12) is a single-valued expression that avoids the multiple-root ambiguity of equation (9). Second, $C2_{100}$ and $G3_{max}$ should both be proportional to the "size" of the battery. Thus, battery "size" may cancel out of equation (13). If this indeed holds true, equation (12) will prove to be a much more useful expression for SOC^- than either equation (9) or equation (11).

Determining SOC from Open-Circuit Voltage

The conventional method for determining state-of-charge of an SLA battery is to measure its open-circuit voltage (OCV). This is an indirect method since OCV is primarily affected by the concentration of SO_4^{2-} ions in the electrolyte; not Pb or $PbSO_4$ molecules on the electrode surfaces. Furthermore, the OCV result is significantly affected by the initial electrolyte acid concentration as well as by "surface charge". Nevertheless, this technique is commonly used.

According to Hammel (8), pp. 15-4 and 15-5, the OCV of a spiral-grid VRLA/AGM battery, such as the one under investigation, varies linearly from 1.97 to 2.17 volts/cell as the SOC varies from 0 to 100%. Hammel

states that the SOC determined from this relationship is accurate to within 20% if the battery has not been charged or discharged within 24 hours; and to within 5% if it has not been used for 5 days.

Figure 16 shows a plot of SOC determined in this manner as a function of Ah removed. One sees that the calculated SOC goes through zero and takes on negative values above 30 amp-hours removed. This corresponds to an error of nearly 40 % at 30 Ah removed and even larger errors at higher values of Ah removed. The battery's having been allowed to "rest" for only 2.5 hours after each discharging before an OCV measurement was made may account for much of this discrepancy. However, Figure 16 graphically illustrates a serious problem with using OCV to determine SOC.

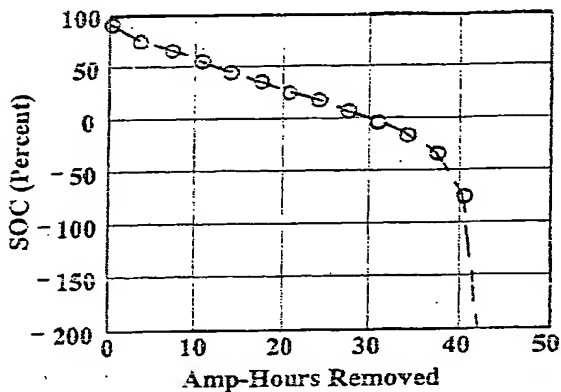


Figure 16. State-of-charge as conventionally determined from open-circuit voltage.

DFIS MEASUREMENTS DURING CHARGING

Figures 17, 18, and 19 display the results of DFIS measurements that had earlier been performed on this same VRLA/AGM battery while it was being charged. The charging experiments began with the battery in an approximate "half-charged" condition. The battery was then charged at constant current until a terminal voltage of 14.4 volts was reached. Three-frequency DFIS measurements were continuously performed as the battery was undergoing charging. Unfortunately, nearly one year intervened between performing these charging measurements and later performing the discharging measurements that yielded the data of Figures 8, 9, and 10. Nevertheless, there is remarkable agreement between these measurements and the theoretical interpretation of the discharging measurements detailed above.

Figure 17 displays measured battery voltage as a function of "Amp-Hours Injected", the opposite of "Amp-Hours Removed". One sees that the battery voltage increases quite linearly to 13.3 volts at approximately 21 Ah injected. It then increases steeply to the cut-off voltage of 14.4 volts at 23.5 Ah injected.

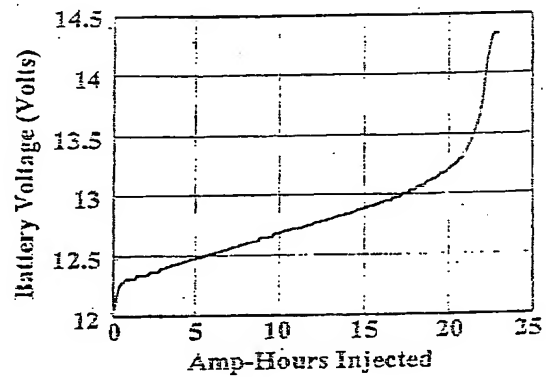


Figure 17. Battery voltage during charging.

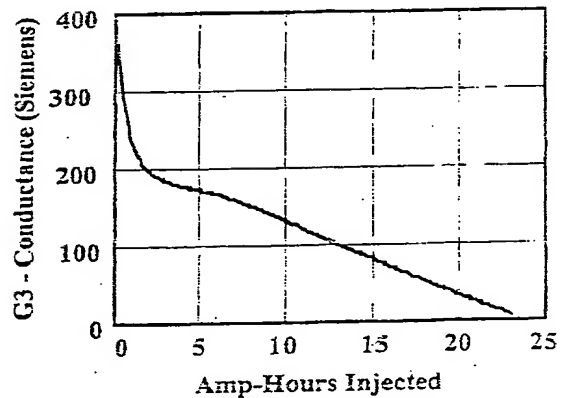


Figure 18. Conductance G3 during charging.

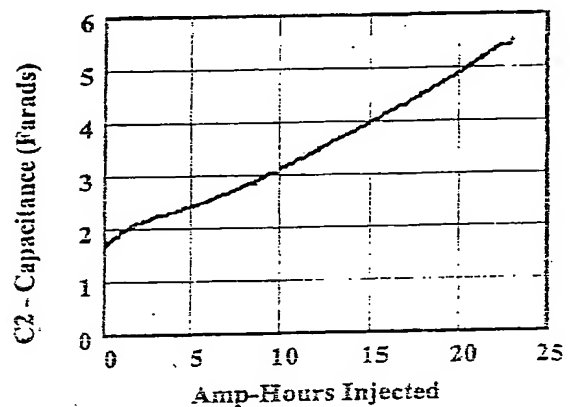


Figure 19. Capacitance C2 during charging.

Figures 18 and 19 display measured conductance G3 and measured capacitance C2, respectively, over this same range of Ah injected. These plots actually represent 170 individual DFIS measurements. Such a large number of measurements would be completely impossible to perform using conventional EIS

methodology. Remarkably little scatter is noted between the individual DFIS measurements.

Figure 18 shows that conductance G_3 decreases with increasing Ah injected and actually reaches zero when 23.5 Ah have been injected. Such behavior is completely consistent with the variation of G_3 with SOC^- predicted by equation (7). According to equation (7), conductance G_3 reaches zero when $(1 - SOC^-) = 0$. This condition indicates that all of the reaction sites on the negative electrodes have been converted from $PbSO_4$ to Pb -- thus leaving no sites available for the "charge" reaction. The observed voltage rise occurs because the electrical energy that had been going into the "charge" reaction must now go into other processes -- such as electrolysis and heating.

Figure 19 shows that capacitance C_2 increases linearly with Ah injected and reaches 5.5 Farads when 23.5 Ah have been injected. Such behavior is completely consistent with equation (10), which reveals that C_2 is proportional to SOC^- . The observed linear increase of C_2 in Figure 19 simply indicates that SOC^- increases linearly with Ah injected -- as is to be expected. The maximum value of C_2 , attained at $SOC^- = 100\%$, is $C_{2,100} = 5.5$ Farads. This number really compares remarkably well with the value $C_{2,100} = (4.176/0.936) = 4.5$ Farads calculated from the DFIS measurements of Figures 8 and 9 performed nearly one year later.

CONCLUSIONS

Discrete Frequency Immittance Spectroscopy (DFIS) has been shown to be a convenient and accurate technique for obtaining meaningful information about real lead-acid batteries. Although DFIS is related to conventional EIS, it is much easier to perform and yields results in far less time. The excellent agreement between theory and experiment attests to the validity of those results.

A number of innovative concepts and new results have been presented. Among them are:

- DFIS measurements on real lead-acid batteries at frequencies above 5 Hz yield information about the electrochemistry of the *negative electrode surfaces*.
- An electrical over-voltage at the external terminals splits into *electrical* and *chemical* components of the *electrochemical overpotential* at the electrode surfaces. These two components add in *series* in the DFIS circuit model.
- Conductance G_3 is associated with the *chemical overpotential* and is directly proportional to the *exchange current* of the negative electrode surfaces.

- Capacitance C_2 is associated with the *electrical overpotential* and is equal to the *ionic double-layer capacitance* of the negative electrode surfaces.
- A *negative electrode state-of-charge* is defined to be equal to the fraction of negative-electrode reaction sites that are occupied by PB molecules.
- This *negative electrode state-of-charge* can be expressed in terms of either G_3 , C_2 , or a combination of G_3 and C_2 .
- The *negative electrode state-of-charge* is less than 100 % when the battery is "fully charged" in the conventional sense.
- Charging the battery until the *negative electrode state-of-charge* reaches 100 % causes G_3 to go to zero and the battery voltage to rise abruptly.

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Applicant : Harvey A. Restaino et al.

Serial No./Patent No.: 10/783,682

Filed/Issued : February 20, 2004

Title/Mark : REPLACEABLE CLAMP FOR ELECTRONIC BATTERY TESTER

1. Supplemental Information Disclosure Statement



Atty/Sec LRT/jme Date: 7/11/05 File No. C382.12-0146

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named
Inventor : Harvey A. Restaino et al.

Group Art Unit: 2838

Appln. No. : 10/783,682

Examiner:

Filed : February 20, 2004


For : REPLACEABLE CLAMP FOR
ELECTRONIC BATTERY TESTER

Docket No. : C382.12-0146

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Atty/Sec LRT/jme Date: 9/24/04 File No. C382.12-0146

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named
Inventor : Harvey A. Restaino et al.

Group Art Unit: 2838

Appln. No. : 10/783,682

Examiner:

Filed : February 20, 2004

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